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VISIBLE LASER RESEARCH  
ONE METER DEVICE

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Interim Technical Report for Period 1 July 1979 to 28 February 1980

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Blue-Green Lasers Deuterium Stimulated Raman Scattering XeF* Hydrogen lambda		
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Stimulated Raman Scattering Experimental Results are discussed. The pump laser was XeF ( $\lambda = 350$ nm) and two step conversion to the blue-green spectral region was accomplished using molecular hydrogen and deuterium. Conversion efficiencies were measured and overall conversion from the UV to the blue-green of >40% is reasonable.		

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## I. INTRODUCTION

There is an ongoing need in a variety of Department of Defense applications for a dependable, efficient, high average power laser operating at ocean water transmitting wavelengths ( $\lambda \cong 470$  nm). It has also been historically true that few lasers operating near this optimal wavelength are available, let alone meet the requirements for efficient, scalable operation. To develop a blue-green laser system, one can either explore new laser concepts leading to direct lasing in this sought-after bandpass or undertake means to shift the output wavelength of existing, proven lasers into the desired regime. One such class of proven lasers are the exciplex lasers which include the rare gas halides, halogens and mercury halides (see Table I). Of these exciplex systems, there are four that have a demonstrated capacity for respectable efficiencies and scalable, high power operation; namely ArF\* (193 nm), KrF\* (249 nm), XeCl\* (308 nm) and XeF\* (351 nm). Of these, we feel XeF\* shows the most promise as a basis for an efficient converter. This is based principally on its suitability for scaling to high power and the fact that the blue-green spectral region can be readily accessed by stimulated Raman output from XeF\* pumped molecular hydrogen.

For any stimulated Raman process phenomenologically, the acceptor molecule (e.g., H<sub>2</sub>) can be thought of as absorbing an incident photon (e.g., XeF\* 351 nm) thereby making a transition to a virtual state and then, with the emission of a Raman photon at longer wavelengths, proceeding to a level near the ground (initial) state (e.g., H<sub>2</sub> ( $v = 1$ )). Stimulated Raman scattering in hydrogen by rare gas halide pumps has been studied at LASL.<sup>(1)</sup> The forward scattering cross section is large and overall energy conversion efficiency of  $\sim 80\%$  has been observed with  $\geq 50\%$  in the 1st Stokes line routinely reported.<sup>(1)</sup>

**TABLE I**  
**A SUMMARY OF EXCIPLEX LASERS AND THEIR LASING WAVELENGTHS**

EXCIPLEX	$\lambda$ (B $\rightarrow$ X) nm
ArF	193
KrCl	222
KrF	249
XeBr	282
XeCl	308
XeF	351 (B $\rightarrow$ X), 500 (C $\rightarrow$ A)
$F_2$	158
FCl	284
$Br_2$	292
$I_2$	342
IF	491
Hgl	440
HgBr	502
HgCl	558

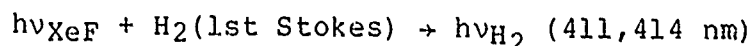
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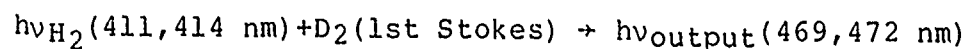
We have been experimentally investigating wavelength conversion of XeF\* to the blue-green using this technique of stimulated Raman scattering involving hydrogen and deuterium.

In particular, our approach involves sequential conversion through two distinct, separate steps involving H<sub>2</sub> and D<sub>2</sub>:

Step 1

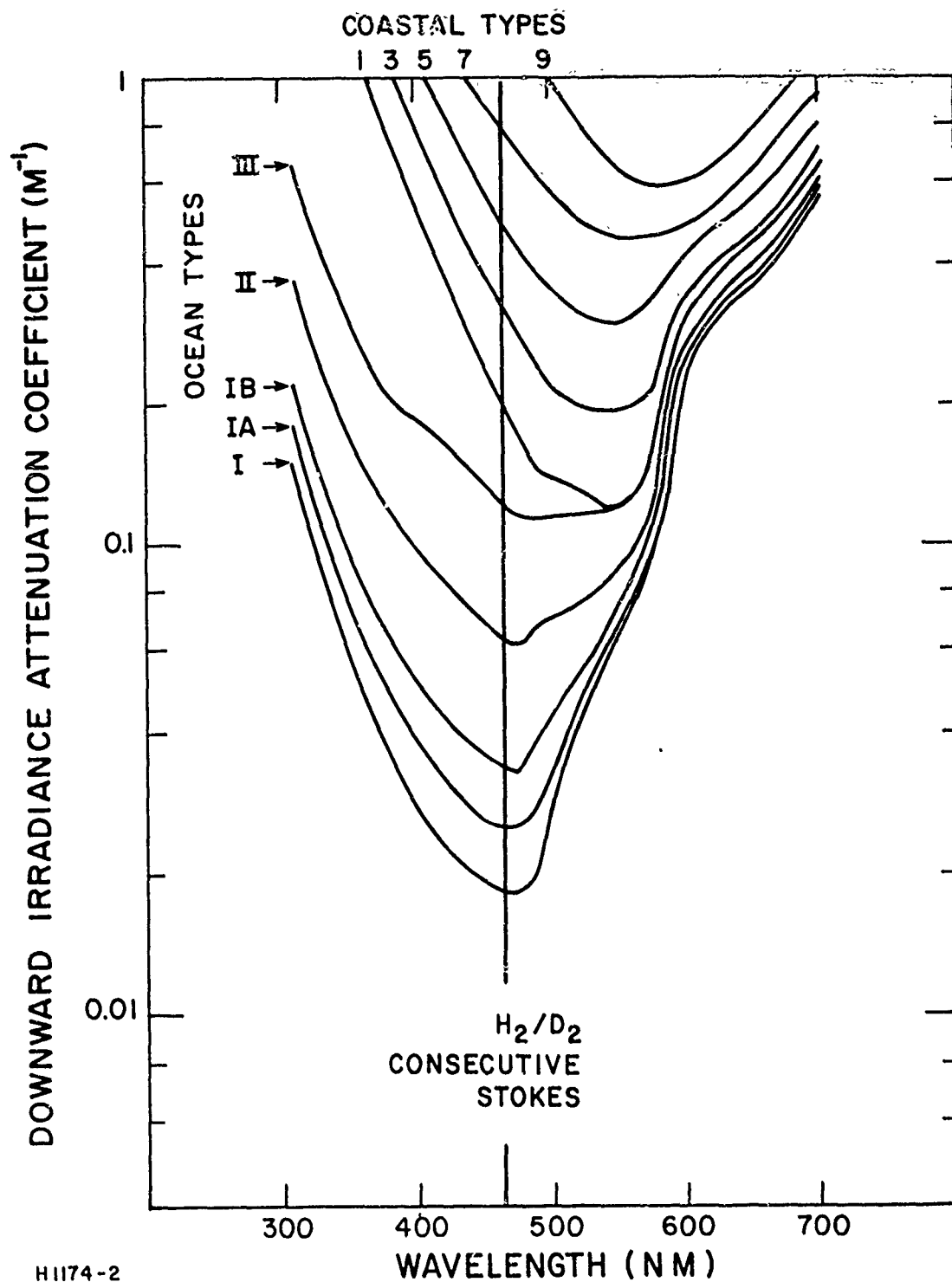


Step 2



The use of two separate cells provides wavelength flexibility through the choice of appropriate Raman scattering media in the various cells. In addition, optimization on first Stokes in both cells, sequentially, each in a single pass, is a straightforward method for minimizing the effects of four-wave, parametric mixing processes which can reduce the desired Stokes output.

The ability of the AERL two-step approach to select a desired wavelength through the choice of the gases contained in the two cells can have significant system advantages, since various ocean water types have different wavelength transmission properties (see Figures 1-5). Figure 1 shows the Jerlov attenuation coefficients as measured through the top 10 m of surface water classified according to ocean types. Figure 2 plots the same data as attenuation of incident light through 100 m of ocean water, assuming the top 10 m are characteristic of the entire optical path length. For types I, II and III water, there is a clear advantage in transmitted signal for wavelengths near 475 nm, and a laser operating at or near this wavelength would be appropriate for all three ocean types. For coastal types, longer wavelength lasers offer some advantage but the extreme signal attenuations involved may suggest that wavelength may not be the single most significant issue.



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Figure 1 Ocean Water Attenuation Coefficients vs Wavelength  
Indicating XeF\* Raman Conversion Wavelength

# OCEAN WATER ATTENUATION TO 100 METERS (JERLOV DATA)

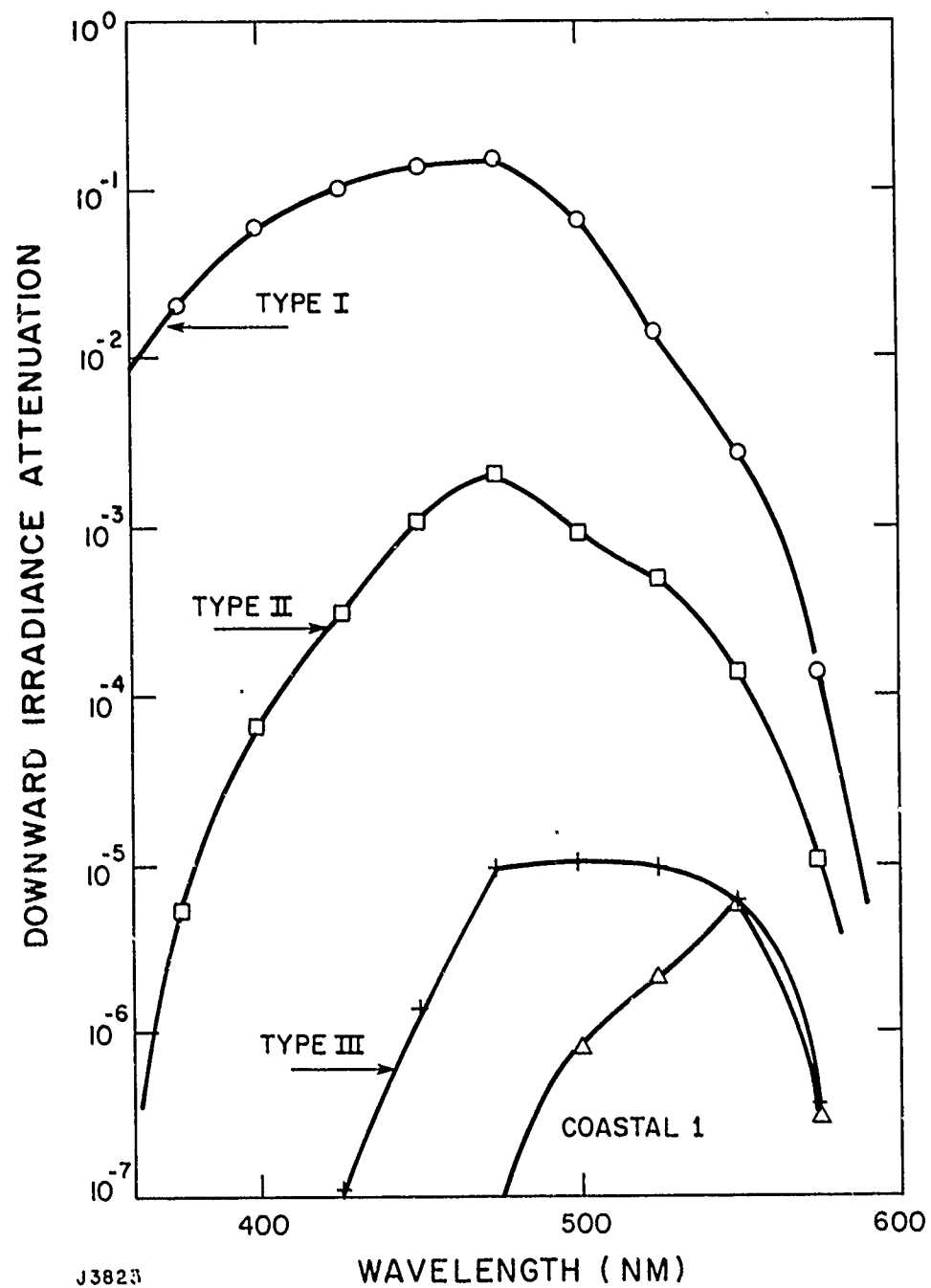


Figure 2 Downward Irradiance Attenuation vs Wavelength

# OCEAN WATER TRANSMISSION TO 100 METERS (JERLOV DATA)

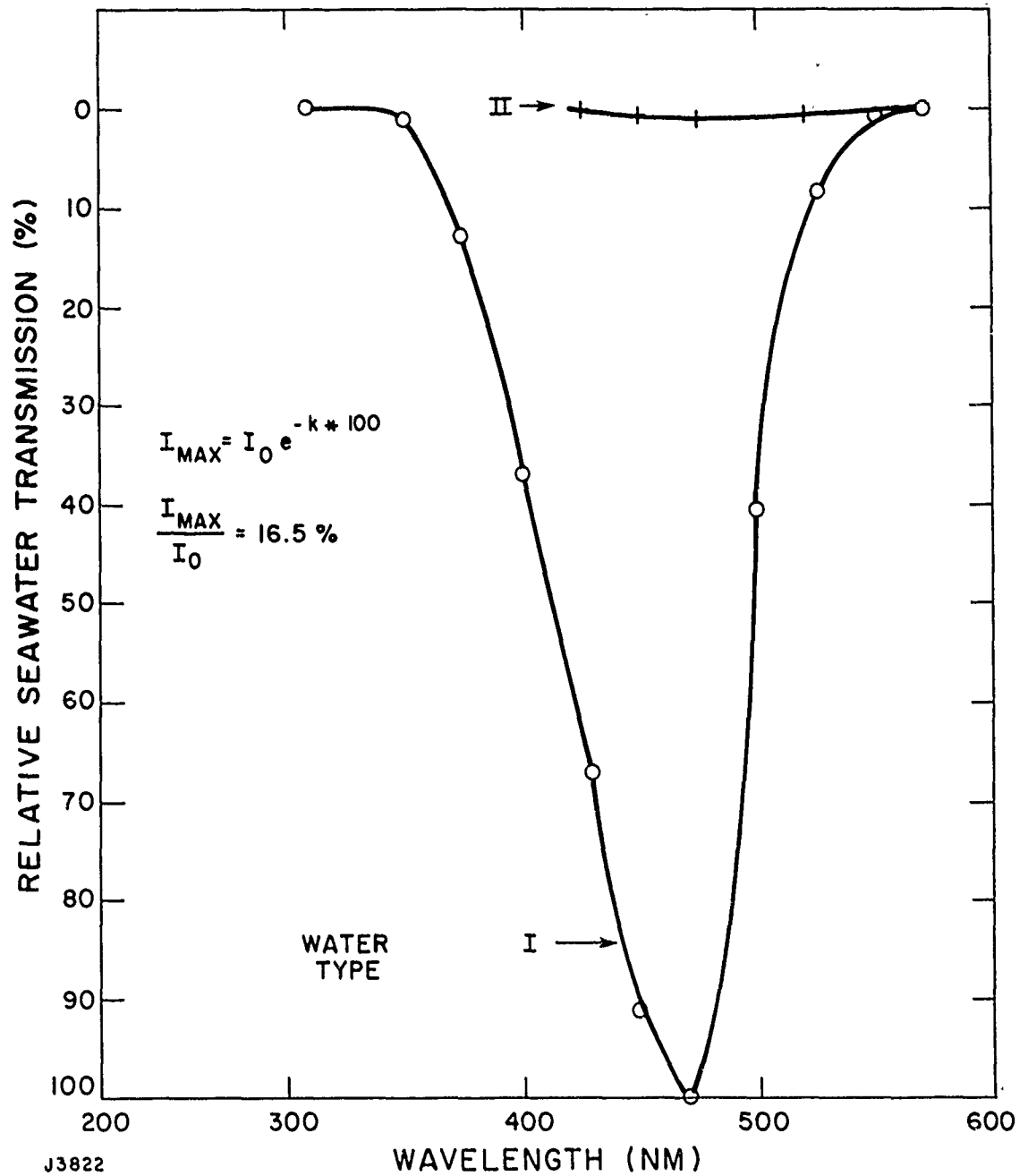


Figure 3 Relative Seawater Transmission vs Wavelength for Water Types I and II

# OCEAN WATER TRANSMISSION TO 100 METERS (JERLOV DATA)

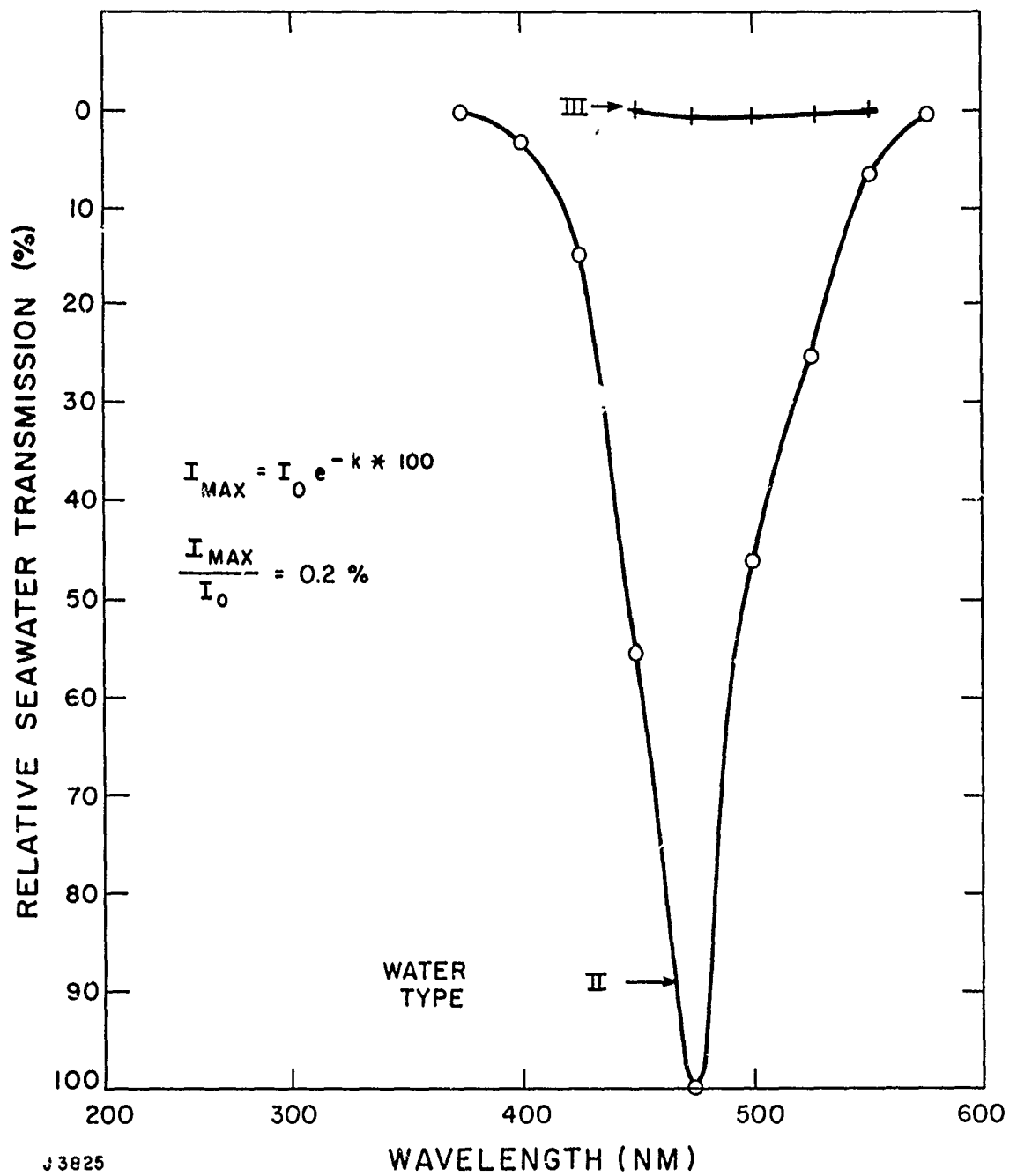


Figure 4 Relative Seawater Transmission vs Wavelength for Water Types II and III

# OCEAN WATER TRANSMISSION TO 100 METERS (JERLOV DATA)

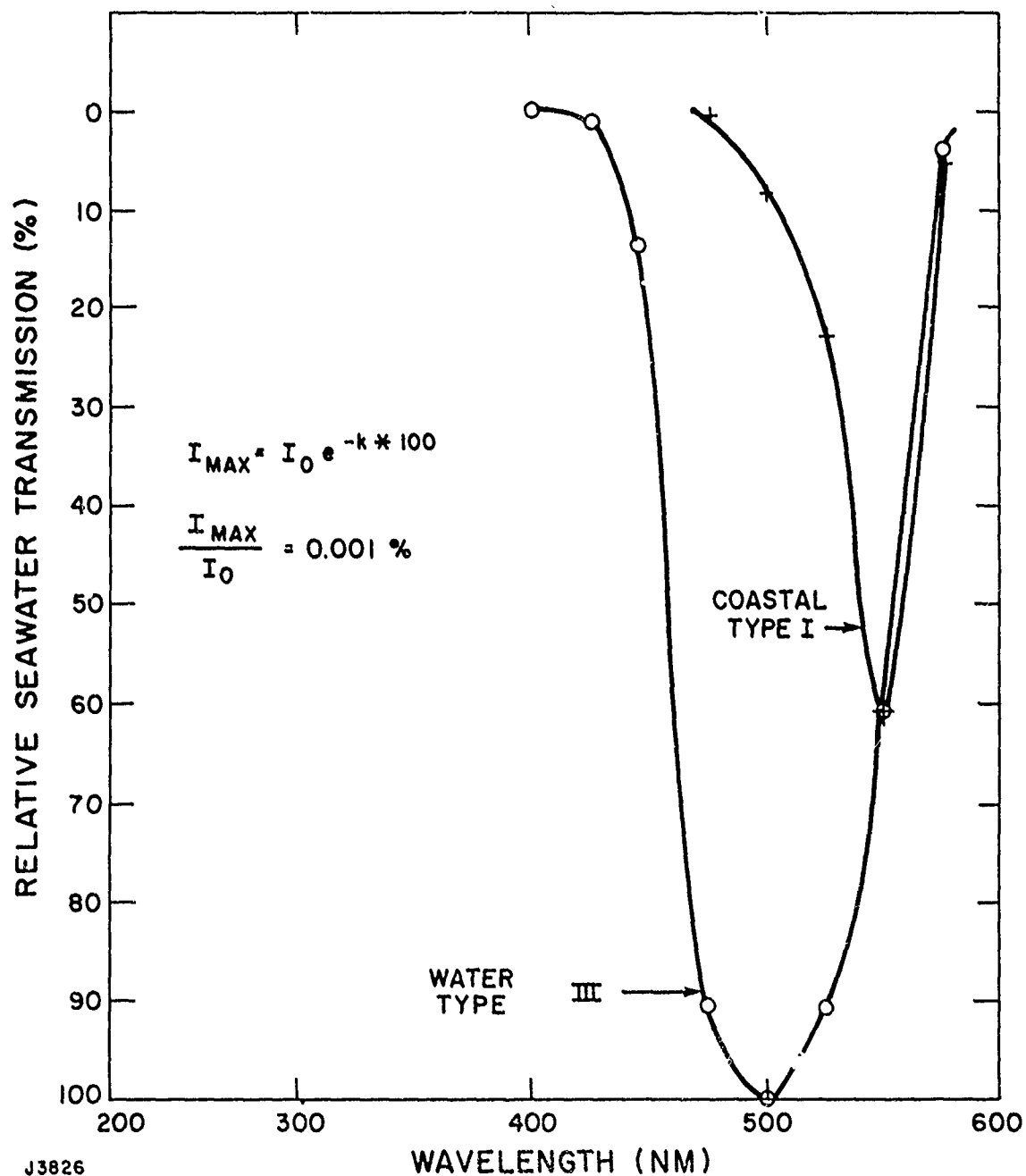


Figure 5 Relative Seawater Transmission vs Wavelength for Water Type III and Coastal Type I

This influence of wavelength is depicted on a relative basis in Figures 3-5. Here the peak transmission is listed for the optimum wavelength along with a plot of the relative wavelength scaling for each ocean type normalized to the peak. For example, Figure 4 shows maximum 100 m transmission through ocean type II is 0.2% at 475 nm. At this same wavelength, type III transmits a very small percentage (see top of Figure 4) and itself is plotted with coastal type I in Figure 5. These graphs, which plot transmission on a linear scale, demonstrate more clearly the significant dependence of the transmitted signal on wavelength.

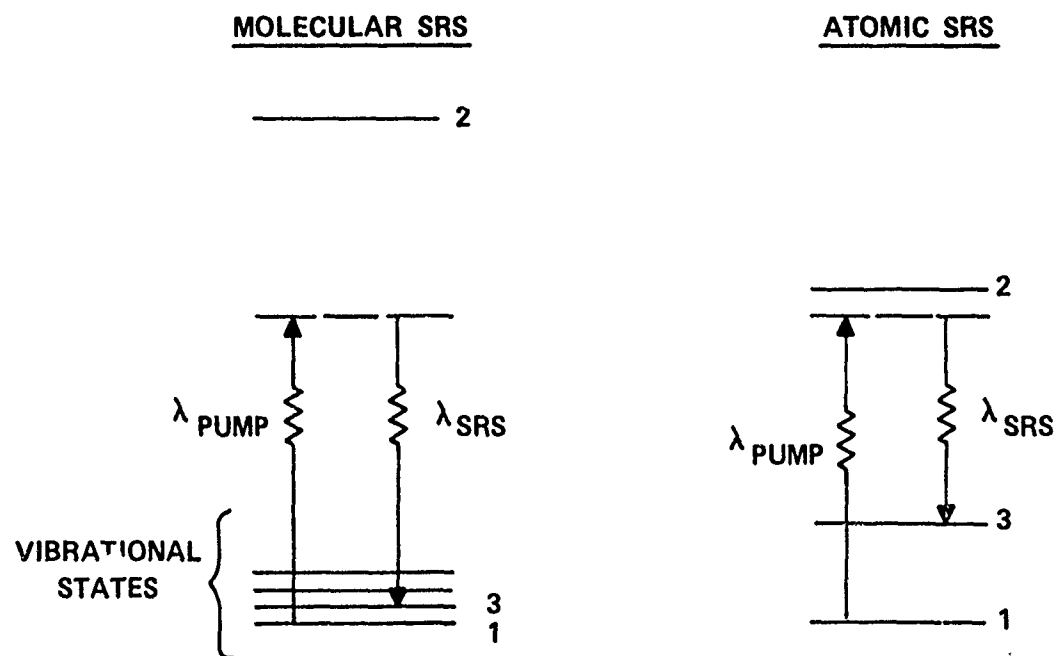
Blue-green lasers operating as near to the ocean transmission window as possible provide the greatest potential signal-to-target, thus reducing the required laser power needed to provide any minimally acceptable receiver signal. The output wavelengths of 469 and 472 nm, corresponding to our approach of two-step conversion with  $D_2$  and  $H_2$  of the two lasing transitions of  $XeF^*$ , are nearly optimum for ocean water transmission based on these Jerlov data. Since high conversion efficiency has been demonstrated in  $H_2$  at AERL with an  $XeF^*$  laser pump, we project that, starting with an  $XeF^*$  overall efficiency of  $\geq 4\%$ , an overall blue-green laser efficiency of  $\geq 1\%$  is attainable.

## II. BACKGROUND

In general, for the stimulated Raman process both atomic and molecular gases can act as the nonlinear medium, and some of the differences between the atomic and molecular cases are illustrated in Figure 6. For any stimulated Raman process, the acceptor atom or molecule can be thought of as absorbing an incident photon (e.g., XeF\* 351 nm) thereby making a transition to a virtual state and then, with the emission of a Raman photon at longer wavelengths, proceeding to a level near the ground (initial) state. For an atomic candidate, the practical constraints involve searching for an atom which has a dipole allowed electronic transition near the pump laser frequency and a corresponding transition from that upper state (2) to a lower state (3) at the sought-after conversion wavelengths. Specifically, for this blue-green mission, workers at the Naval Research Laboratory (Ref. 2) have reported high conversion efficiency of XeCl (308 nm) to 459 nm using atomic lead as the scattering candidate. These atomic systems do, however, have the singular disadvantage of requiring high-temperature production techniques (heat pipes) which may prove technologically difficult at the very high average powers under consideration for actual systems applications. In addition, the typical achievable metal atom density requires the use of the metal atom more than once during the laser pulse (recycling) or large volume sources, since one must have at least as many metal atoms as photons for high conversion efficiency. For molecular Raman scattering (see Figure 6), the process is generally the same as in the atomic case but is much more non-resonant. Since there is no resonant enhancement of the cross section, large densities are usually required to produce reasonable gain for readily available pump lasers.

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Figure 6 Optical Conversion by Stimulated Raman Scattering

### III. RESULTS

#### A. SHORT-PULSE LASER RESULTS

For the past eight months, we have been investigating efficient generation of Stokes shifted radiation using a commercial exciplex laser as the pump (Lumonics, TE-261-2) and molecular  $H_2$  and  $D_2$  as the Raman scattering media. General laser specifications as detailed by the manufacturer were found to be achievable with our particular device (e.g., it produced  $\sim 75$  mJ of energy per pulse with  $XeF^*$  and  $\sim 190$  mJ for  $KrF^*$  with the cavity optics provided). The beam quality associated with these achievable energies was insufficient to provide focussed intensity-length products to achieve laser action in the Raman gas. We, therefore, altered the cavity optics to include Brewster windows and an unstable cavity configuration of the type described by Barker and Loree (Ref. 3). This cavity produced  $\lesssim 20$  mJ of  $XeF$  laser energy in a pulse (FWHM) of  $\simeq 6$  ns with a beam waist 10 times the diffraction limited value. The power density achievable was measured at the focus by recording the energy transmitted through various diameter pinholes. The beam waist at the focus of a 50 cm fl lens was found to be near  $2.5 \times 10^{-2}$  cm in diameter with an energy of 10 mJ. This, with the 6 ns pulselength, corresponds to an achievable power density of  $\sim 3 \times 10^9$  W/cm<sup>2</sup> at the focus.

Efficient conversion however relies on an intensity-density-length product well in excess of threshold to achieve optimum conversion of  $XeF$  pump radiation to first Stokes. To measure efficiency and to parameterize the Raman process, the experimental setup shown in Figure 7 was used. Here the output of the Lumonics was focussed through a pinhole to 1) characterize the input beam, and 2) eliminate that portion of the pump with poor beam quality. In this arrangement, a second lens refocusses the beam into the center of our high-pressure Raman cell.

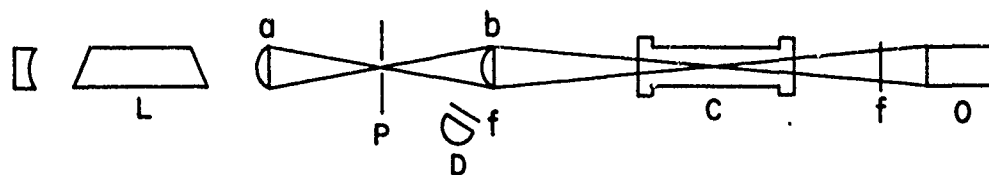


Figure 7 Schematic of Experimental Setup where L is the Lumonics Laser, a and b are 50 cm fL lens, C is the High-Pressure Cell, D is a Photodiode, F are Filters and O is a Spectral Monitoring or Energy Measuring Device

The Raman cells were constructed from available stainless steel shock tube sections about 40.6 cm long with an inner diameter of 3.8 cm and a wall thickness near 1.3 cm. The polished Ultrasil quartz windows were near 6 cm in diameter and ~ 2.5 cm thick. The windows were supported by O-rings on the high-pressure side and by gaskets on the low-pressure side. The internal volume of these cells was near 465 cm<sup>3</sup>.

Pulse energies in these experiments were measured with a Scientech Model 362 energy/power meter. Pulse shapes were measured by photodiodes (ITT, type S-5 or Hamamatsu, type S-4). The outputs from the photodiodes were recorded by a Tektronics Model 7844 dual beam oscilloscope equipped with a C-51 oscilloscope camera.

Spectra were analyzed with either a 3/4 m monochromator, 1 m spectrograph, or an optical multichannel analyzer (Tracor Northern, Model TN-1710, equipped with a Diode Array Rapid Scan Spectrometer).

Using this arrangement and these various diagnostics, the effects of laser intensity, focal length, and H<sub>2</sub> pressure on the stimulated Raman scattering threshold, gain, Stokes production and conversion efficiency were investigated.

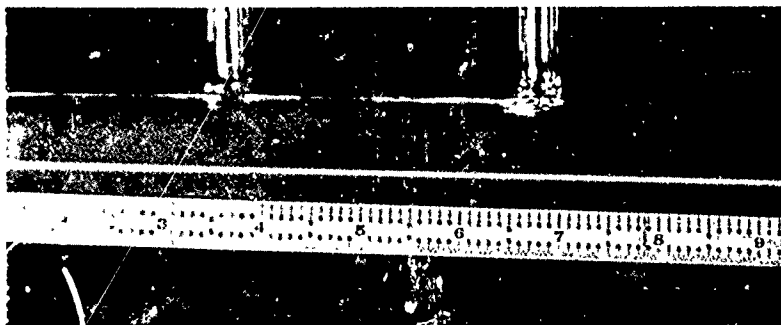
The pump laser intensity (at fixed focal length) was varied in two ways: by repeated firings to degrade the mix from optimum or by lowering the laser charging voltage. Using either technique, the subsequent results were identical, so the more convenient technique of varying the charging voltage was usually employed. Since these experiments were all carried out with focussed geometry, a related factor to the intensity achieved (W/cm<sup>2</sup>) is the length over which the intensity was high. A visual perspective of the various focal arrangements used is shown in Figure 8. These pictures were obtained by open shutter exposures of the focussed laser light into a glass



20 cm FOCUS



50 cm FOCUS



135 cm FOCUS

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Figure 8 Focal Length Profiles

cell containing a dilute solution of Rhodamine 6G. The focal length of  $\sim 135$  cm geometry provided a beam waist of about 0.05 cm in diameter. Figure 9 shows a plot of the energy through this size pinhole vs distance from focus. The distance between half-power points (or the effective Rayleigh range) is about 8 cm. Since the active length of the high-pressure cell is over 40 cm, there was no problem anticipated with gain clipping and no effect on the Raman output was observed when a Raman cell of  $\sim 90$  cm was used.

For this experimental setup (at a maximum laser output corresponding to near  $4.5 \times 10^8$  W/cm<sup>2</sup> for this relatively soft focal geometry), the dependence on forward scattered Stokes generation on H<sub>2</sub> pressure was measured (see Figure 10), using the optical multichannel analyzer (note that the intensity vs wavelength calibration has not been measured). The first Stokes component appears to optimize at pressures above 15-20 atms ( $\sim 250$  psi) whereas the second Stokes appears to peak near 25-30 atm and then decrease at higher pressures. These pressure scaling results are in agreement with the KrF/H<sub>2</sub> experiments of Loree (Ref. 1) and the gain saturation reported by Bloembergen (Ref. 4).

By now keeping the focal length and H<sub>2</sub> pressure fixed, variations in Stokes output were measured as a function of XeF pump input, (see Figure 11). With data similar to these, it is possible to calculate the small-signal gain for this single step process. From our measurements, we found threshold to be near 2.2 mJ. The pulsewidth was 6 ns and the length over which significant conversion occurred was near 8 cm with a beam waist at the center of the focal region of  $\sim 0.05$  cm. Since it is commonly regarded that a gain times length  $\sim 30$  is necessary to achieve lasing from noise (i.e., spontaneous Raman scattering), we can calculate the small-signal gain,  $g$ , as follows:

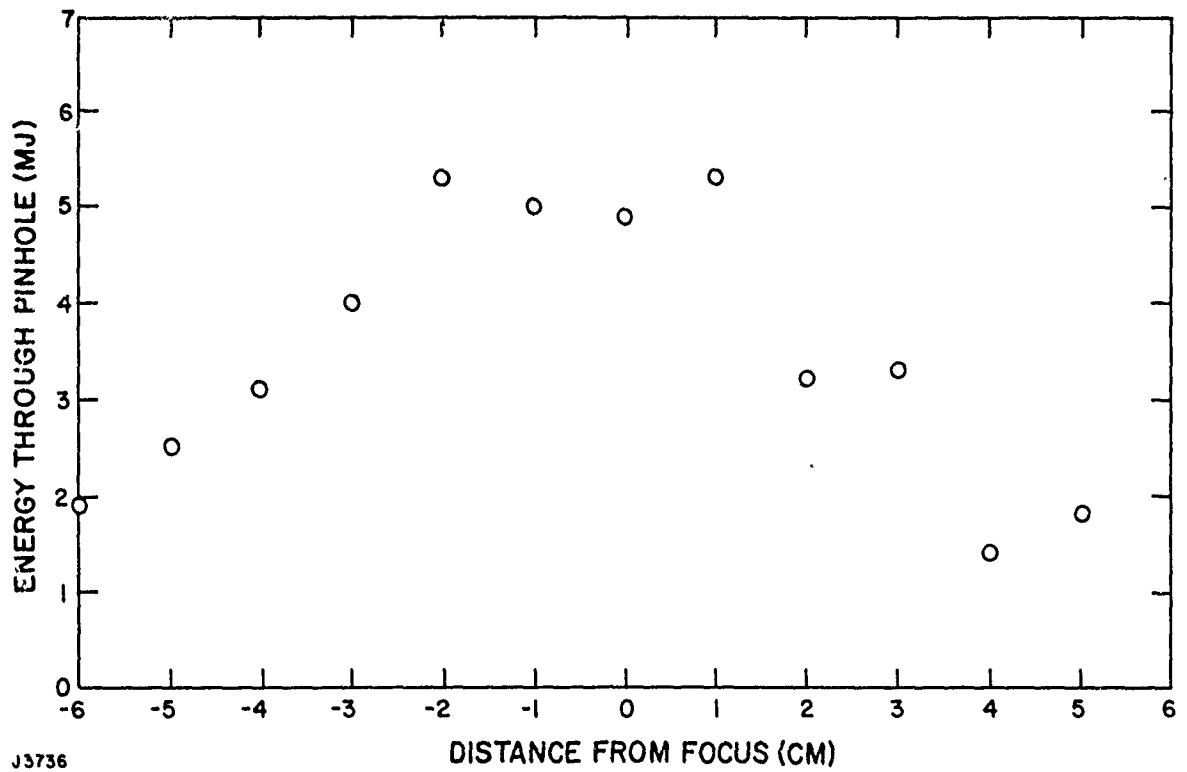


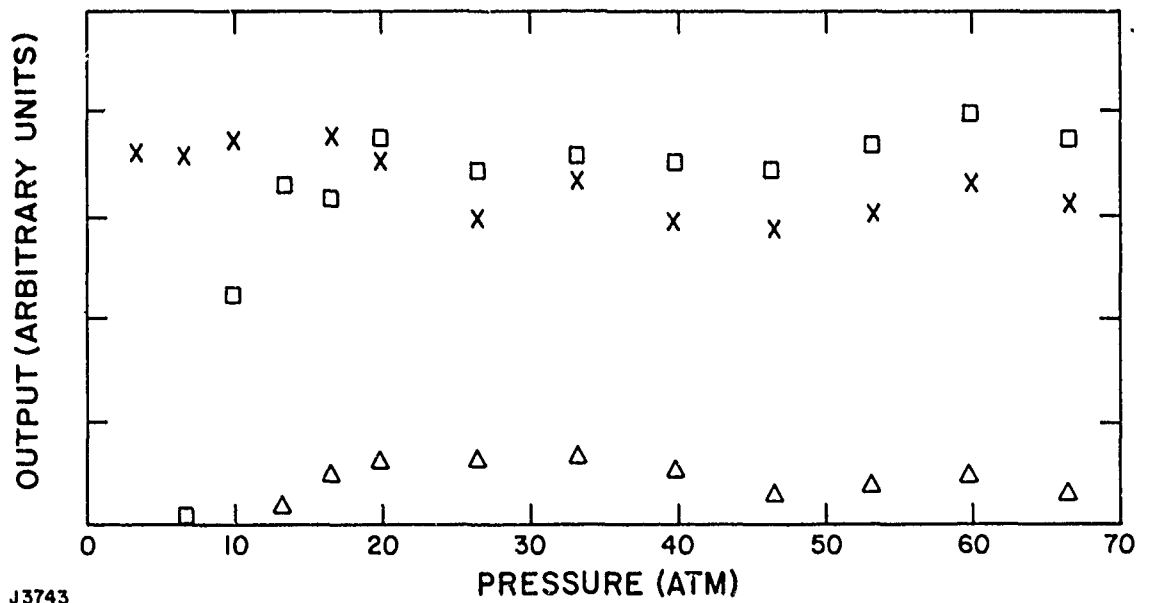
Figure 9 Energy Measured Through a 0.05 cm Pinhole vs Distance from Focus

# H<sub>2</sub> PRESSURE VARIATION (Xe F)

X-DEPLETED PUMP

□-1st STOKES

△-2nd STOKES



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Figure 10 Various Laser Output Photons vs Total H<sub>2</sub> Pressure



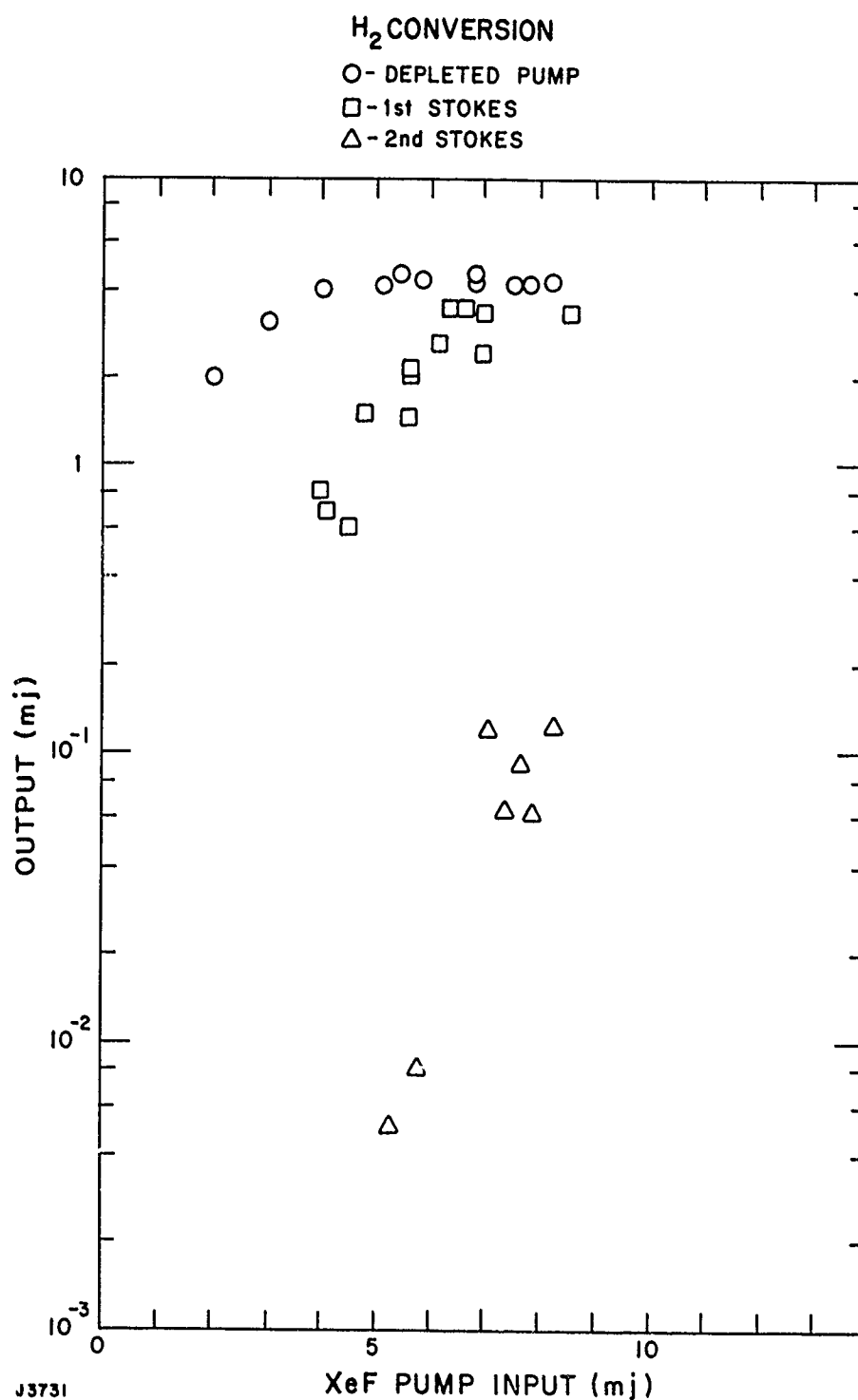


Figure 11 Optimized XeF Conversion to First Stokes vs Input Energy

$$g I_{Th} L \sim 30$$

where

$$I_{Th} = \frac{2.2 \times 10^{-3} \text{ J}}{6 \times 10^{-9} \text{ s} \times \pi (0.025 \text{ cm})^2} = 1.9 \times 10^8 \frac{\text{W}}{\text{cm}^2},$$

and  $L = 8 \text{ cm}$

$$\therefore g \lesssim \frac{30}{I_{Th} L} = \frac{30}{1.9 \times 10^8 \times 8} = 2 \times 10^{-8} \text{ cm/W}$$

This value is about a factor of 5 larger than the value deduced from wavelength scaling of that reported by Bloembergen (Ref. 4) at Ruby wavelengths (see Figure 12). Since the theoretical wavelength scaling for the SRS cross section is fairly straightforward, our results suggest that Bloembergen's reported value is too low. For this blue-green application, our measured small signal gain suggests that this overall Raman approach is relatively easier to accomplish than we had projected in our proposal last year.

The input energy for efficient conversion was also determined from these experiments and the intensity length product needed to accomplish significant conversion was found to be near  $3.8 \times 10^9 \text{ W/cm}$ . This suggests that an XeF\* laser having output near  $10 \text{ J/cm}^2$  in a  $1 \mu\text{s}$  pulse would require (for similar  $\text{H}_2$  densities) a path length of 3.8 m for efficient conversion to first Stokes to occur--a very practical value.

These calculations do represent approximations however, since, for example, the laser output is in two separate lines and the measured threshold energy, beam waist, conversion length, etc. introduce some additional degree of uncertainty. Further measurements to better establish these values are in progress.

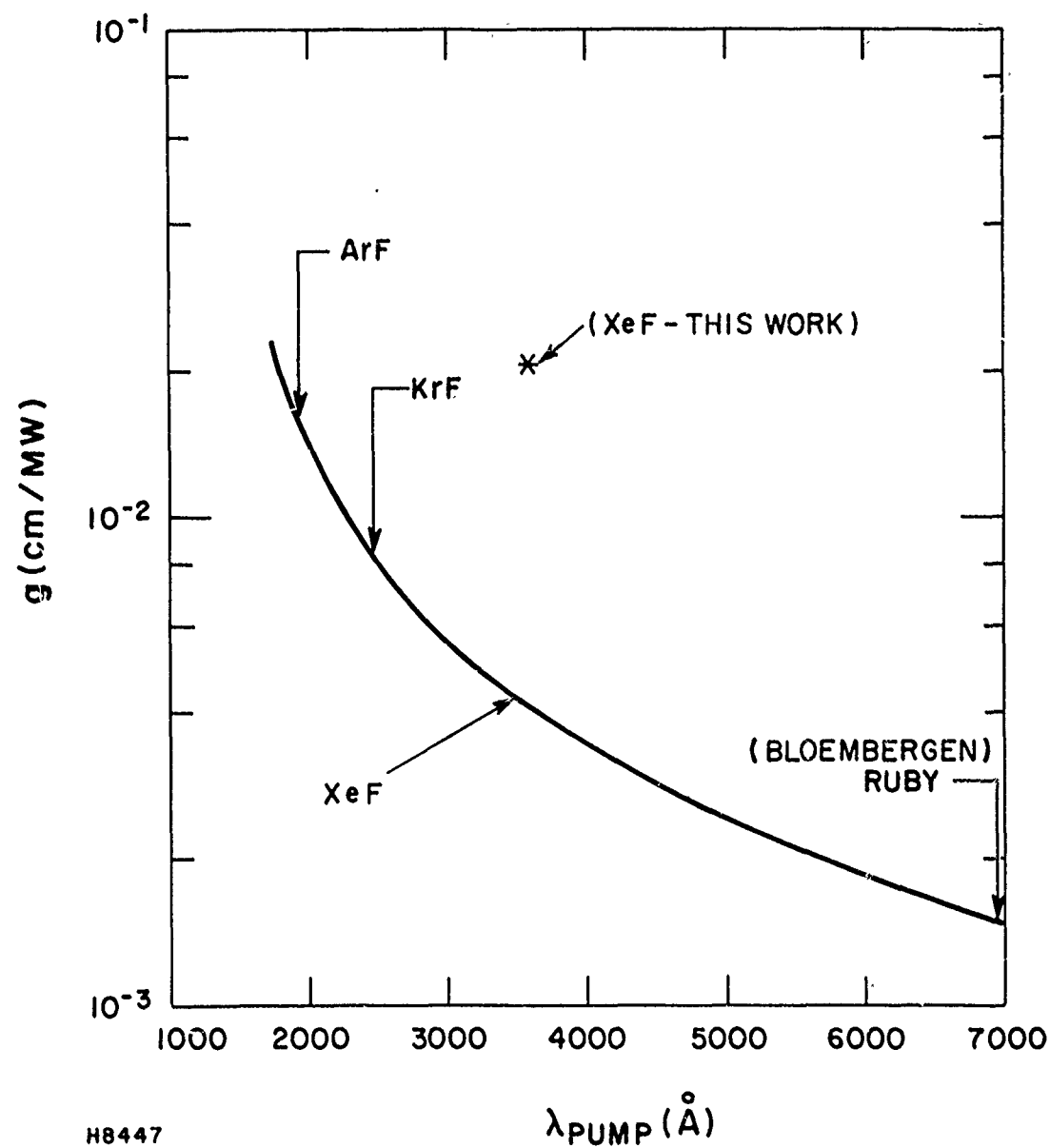
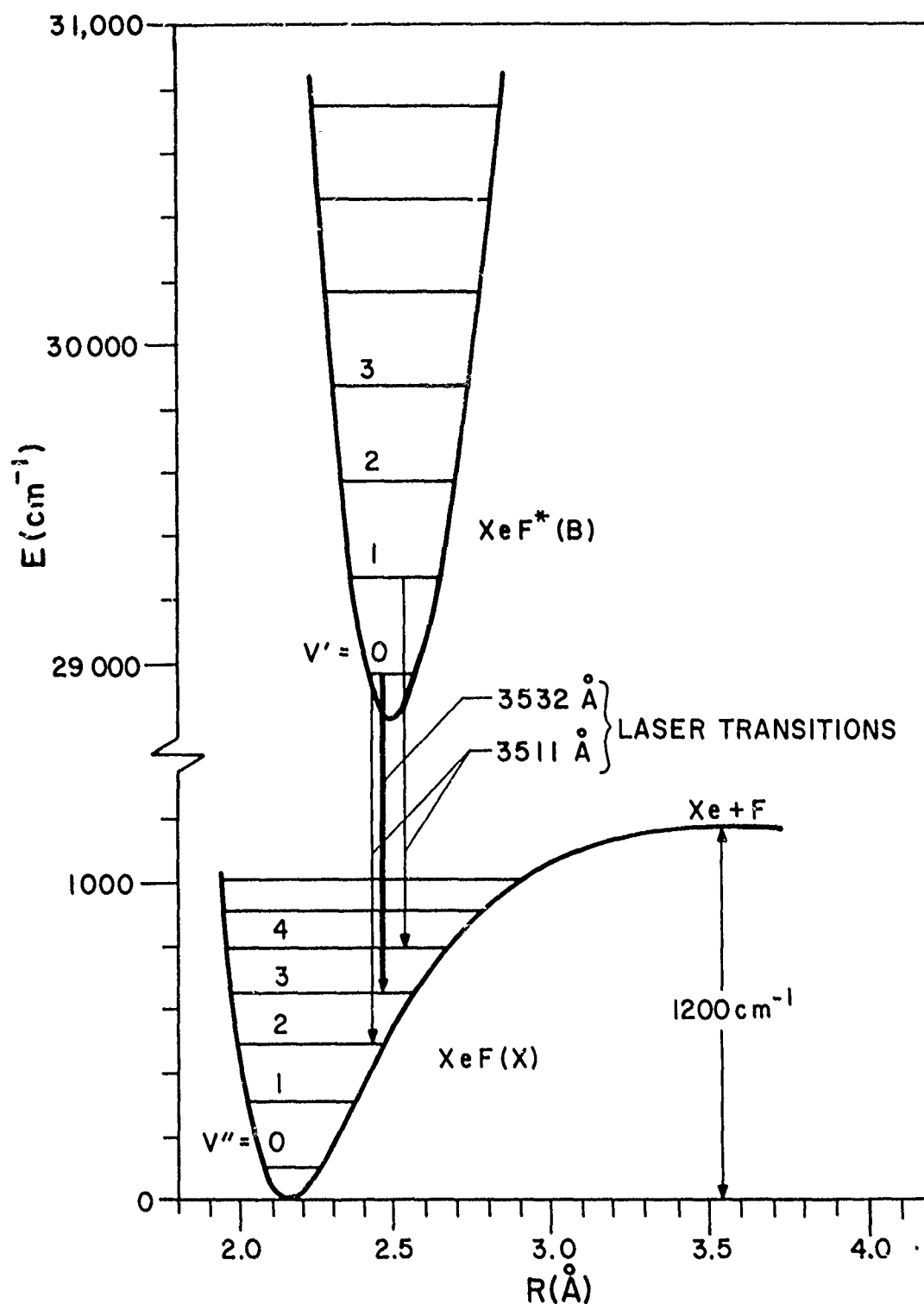


Figure 12 First Stokes Gain for Forward SRS in H<sub>2</sub>. Solid line is wavelength-scaled calculation normalized to Bloembergen's result with ruby; \* indicates the value deduced from this work.

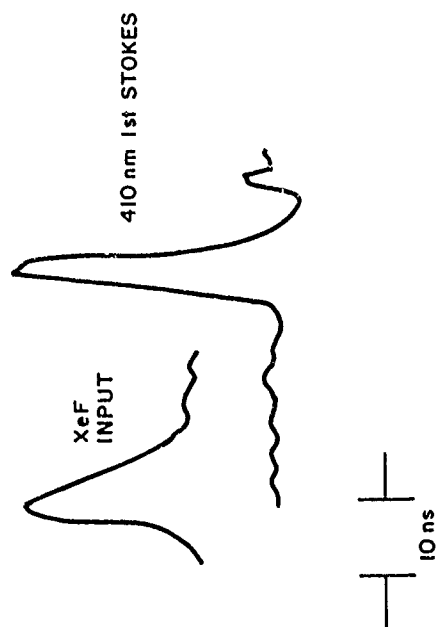
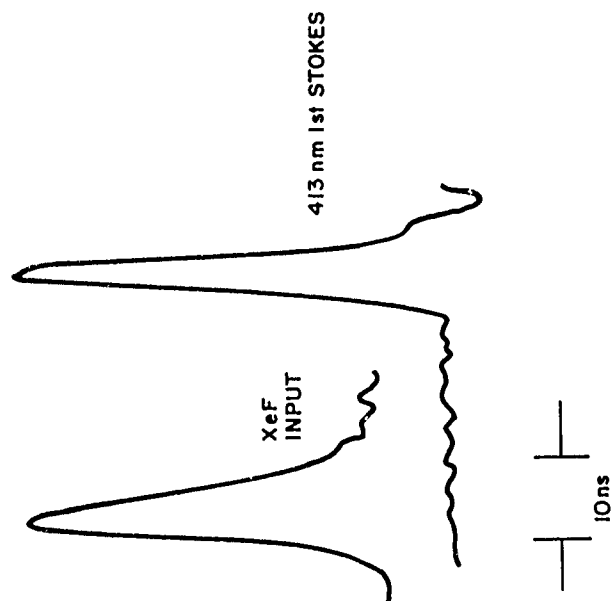
As mentioned above, this pump laser does produce output at both 3511 Å and 3532 Å (see Figure 13) with ~80% at 3511 Å. The total temporal pulsewidth was measured to be 6 ns (FWHM). When these spectral lines were observed independently with a monochromator, their individual pulsewidths were still near 6 ns but the 353 nm line was delayed slightly from the 351 nm line. This observation is consistent with the observations of others of higher gain in the 351 nm band at room temperature (Ref. 5). Pulse shape measurements of the first Stokes pulses were initially performed with the experimental setup using the 50 cm focus (see Figure 8) at pressures of 10 atm. Characteristic pulse shapes are shown in Figure 14. The top traces show the input laser pulse with the 351, 353 nm XeF lines unresolved. The bottom traces are of the corresponding 410, 413 nm first Stokes lines. The temporal widths of these Stokes-shifted lines are ~4 ns. Similar effects were observed for the softer focus. Measurements of the pulse shape of the second Stokes component from a single cell did not show any further pulse distortion, i.e., they had essentially the same 4 ns pulsewidth as the first Stokes.

All of these data and the above discussion allows us to summarize the results of our single pass, single cell, XeF\* pumped H<sub>2</sub> conversion efficiencies for optimized experimental conditions. Conversion efficiencies were calculated by measuring the output energy from the Raman cell through a series of spectral filters, then adjusting the recorded energy for the known transmission of the filters and finally ratioing it to that measured through an empty cell. A photon conversion efficiency of 100% is usually not achievable in these non-resonant molecular Raman systems because as the first Stokes radiation approaches high intensities, it itself acts as a pump and is converted to higher order Stokes emission. Table II contains a summary of the typical output and conversions observed. The



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Figure 13 XeF Lasing Transitions



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Figure 14 Temporal Characteristics of First Stokes ( $H_2$ ) Spectral Lines Using the Short Focal Geometry

TABLE II  
SUMMARY OF SINGLE CELL XeF-PUMPED H<sub>2</sub> SRS CONVERSION EFFICIENCIES

<u>Pump</u>	<u>Focus</u>	<u>E<sub>in</sub> (mJ)</u>	<u>E<sub>out</sub> (mJ)</u>	<u>Energy Eff (%)</u>	<u>Photon Eff</u>	<u>Power Eff</u>	<u>Power Photon Eff</u>
XeF	137 cm	9.6	4.2 S <sub>1</sub>	44	61	66	76
			~ 0 S <sub>2</sub>				

power conversion efficiency is the relevant value, since the energy efficiency should approach the power efficiency for sufficiently long pulselengths. These reported efficiencies did vary somewhat from experiment to experiment, however, the values presented here were observed routinely. (The actual highest efficiency for conversion to  $S_1$  with  $H_2$  in a single experimental observation was 57% energy and 86% power conversion efficiency.)

Other stimulated Raman conversion efficiencies for  $H_2$  reported in the literature include 20% photon efficiency into  $S_1$  pumped by a 1.06  $\mu$  laser by A. Grasiuk (Ref. 6), while Komine and Stappaerts (Ref. 7), report power photon conversion efficiencies of near 50% into either  $S_1$  or  $S_2$ , when  $H_2$  is pumped by a tripled Nd:Yag laser at 355 nm with an oscillator amplifier configuration. Loree, et al. (Ref. 1), have also reported up to 70% energy depletion into various Stokes orders for KrF pumped  $H_2$ .

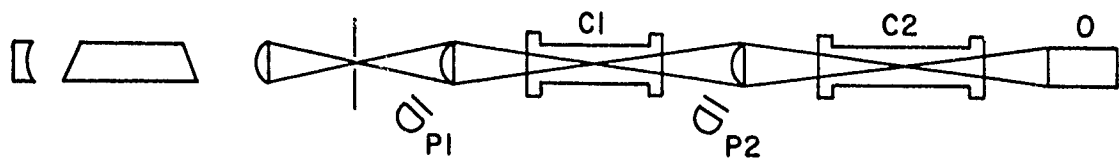
All of these experiments suggest efficient conversion to longer wavelengths via stimulated Raman scattering in a single pass is readily achievable.

The next topic that we addressed in these short pulse, low energy experiments was the second step in the two-step process; namely, investigation of the conversion properties of the emerging  $S_1$  from cell one, subsequently acting as a pump, to produce its  $S_1$  in cell two. (See Figure 15.)

The first of these experiments consisted of two-step shifting through two cells filled with hydrogen, i.e.,  $XeF \rightarrow H_2 \rightarrow H_2$ . The first cell was optimized for maximum conversion of  $XeF^*$  to first Stokes,  $S_1$ , at 410, 413 nm. These emerging laser photons were separated from the depleted pump via a dielectric coated mirror. The depleted  $XeF^*$  pump was thus rejected, while the first Stokes beam from cell one was focussed into the second cell, also optimized to convert into its own first Stokes at 495 nm and 499 nm.



## 2 CELL EXPERIMENTS



J3735

Figure 15 Schematic of Experimental Setup where  $C_1$  and  $C_2$  are the Two High-Pressure Cells and  $P_1$ ,  $P_2$  the Photodiodes for Monitoring the Inputs to each Cell Respectively

Experimentally, the same conversion efficiencies observed through the first cell were not observed in the second step. This was essentially due to the limitations in  $S_1$  energies (acting as the pump) which could be delivered into the second cell through the various optical components (mirror, lens, window, etc.). We were apparently just over threshold in pump laser intensity for the second-step SRS process.

Specifically, the XeF pump laser was focussed into the first cell with a focal length of about 135 cm and the cell was maintained at pressures  $> 40$  atm to insure good conversion. The output light (depleted XeF\* and  $S_1$  radiation) resulting from this optimized configuration was collected and then refocussed into a second cell. A dielectric coated mirror was usually employed, behind the first cell, to reflect ( $> 99.9\%$ ) of the depleted pump yet partially transmit ( $\sim 50\%$ ) the first Stokes radiation. The waist diameter and conversion length of this  $S_1$  radiation was measured as before (see Figure 16). From this figure, the effective gain length is near 5 cm and the power density at the focus is estimated as follows:

0.05 cm pinhole

$$\frac{1.5 \times 10^{-3} \text{ J}}{4 \times 10^{-9} \text{ s}} \frac{4}{\pi (0.05)^2 \text{ cm}^2} = 1.9 \times 10^8 \frac{\text{W}}{\text{cm}^2}$$

0.04 cm pinhole

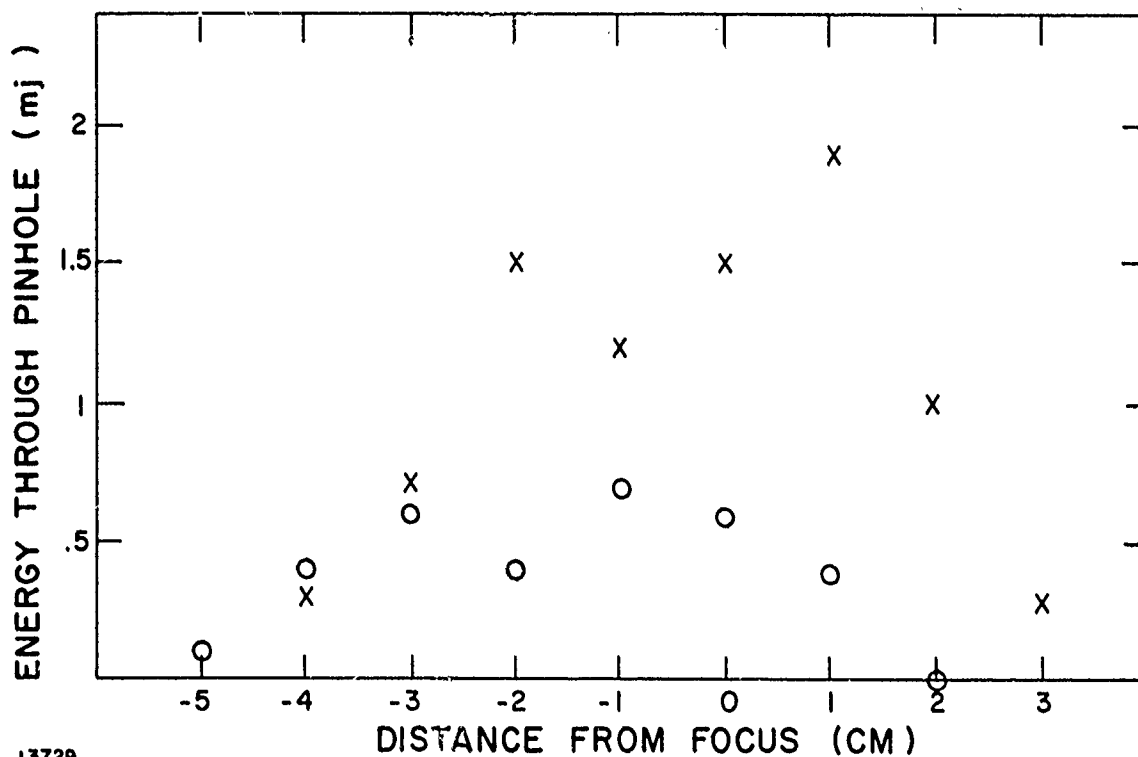
$$\frac{0.7 \times 10^{-3} \text{ J}}{4 \times 10^{-9} \text{ s}} \frac{4}{\pi (0.04)^2 \text{ cm}^2} = 1.4 \times 10^8 \frac{\text{W}}{\text{cm}^2}$$

This represents a lower limit for the actual power density of this first Stokes laser light in that  $\geq 50\%$  of the power was lost in transmission through the optical components. When these are taken into account, the measured power density of the first Stokes compares favorably with the XeF pump ( $4.5 \times 10^8 \text{ W/cm}^2$ ) and indicates that the beam quality of the emerging Raman Stokes shifted light is comparable to the pump.

1<sup>st</sup> STOKES FOCUS AT CELL 2 - H<sub>2</sub>

X - .05 CM DIA PINHOLE

O - .04 CM PINHOLE



J3729

Figure 16 Energy Measured Through 0.05 or 0.04 cm Diameter Pinholes vs Distance from Focus for S<sub>1</sub> Radiant

The variation of the conversion features with the pressure in the second cell was qualitatively similar to that measured in the single cell experiments. Figure 17 shows a plot of the output as measured with the OMA when the pressure in the second cell was varied. (These data are not directly comparable in that the sensitivity of the OMA is greater at 500 nm than at 400 nm but the degree is unknown.) The points at "zero pressure" indicate that the input consisted of both  $S_1$  and  $S_2$ .

Threshold in the second cell was measured by monitoring the first Stokes input with a photodiode and the second Stokes output with another photodiode. The results are summarized in Figure 18. In these experiments, the first Stokes, acting as a pump in the second cell, is probably not far above threshold.

The intensity-length product for conversion of  $\text{XeF}^*/\text{H}_2/\text{H}_2$  can be calculated from these second-cell experiments. The threshold energy was near 1 mJ. The pulselength was 4 ns with a beam waist of 0.05 cm and a conversion length  $\sim 5$  cm at the focus of the second cell. The intensity-length product is, therefore, near  $6 \times 10^8$  W/cm which is considerably less (even after wavelength scaling is accounted for in the calculation) than the value calculated from the single-cell experiments. This lowered (apparent) threshold is caused by the presence of a small amount of second Stokes radiation emitted from cell 1 and entering into cell 2. This small amount of second Stokes can act as the initial input for the conversion process and would require less amplification (i.e., a total gain significantly  $< 30$ ) for stimulated Raman scattering to occur.

The temporal features of the two-step conversion are depicted in Figure 19. This figure shows the XeF pump, the first Stokes output of cell 1 and the second Stokes output of cell 2, when both cells were filled with  $\text{H}_2$ . The pulsedwidth goes from 6 ns for the XeF laser to 4 ns for  $S_1$  to 2.5 ns (FWHM) for  $S_2$ , although the general pulse shapes remained the same.

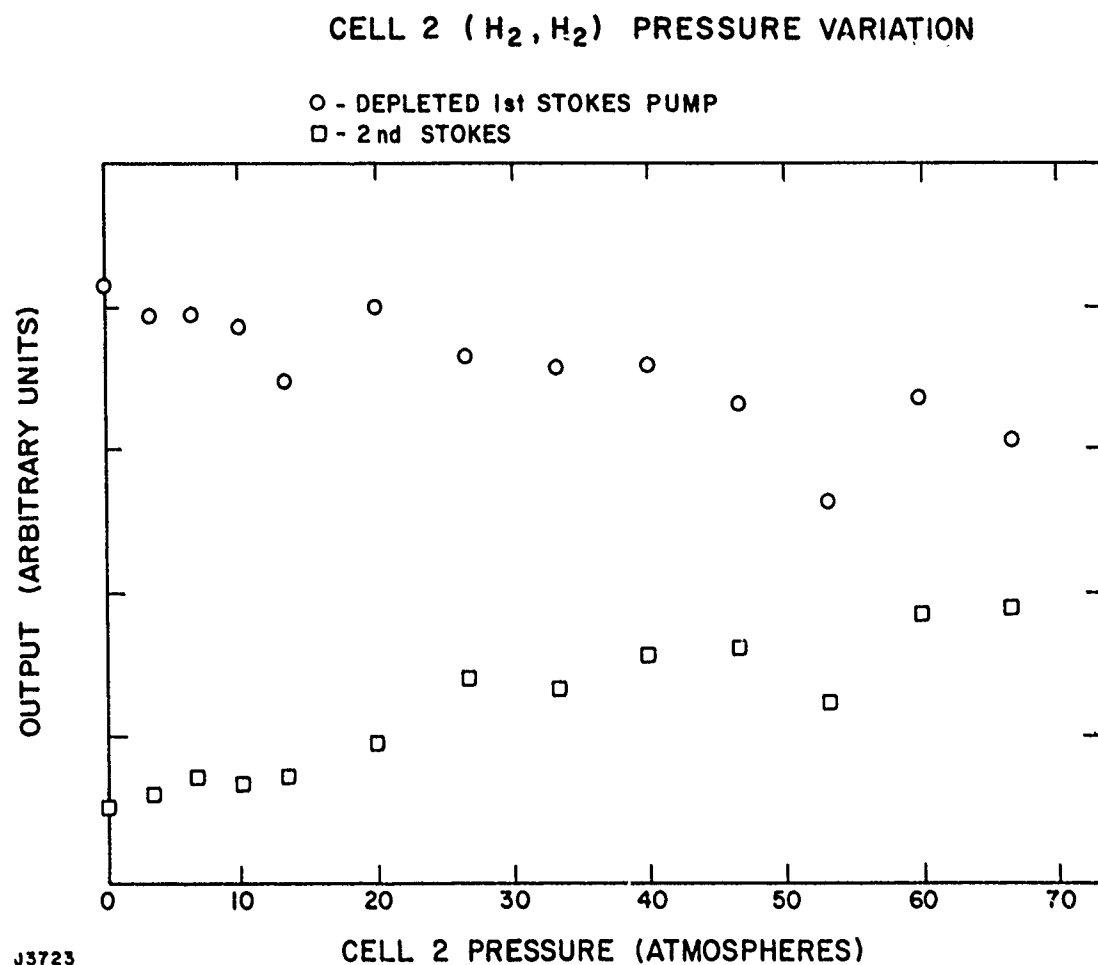
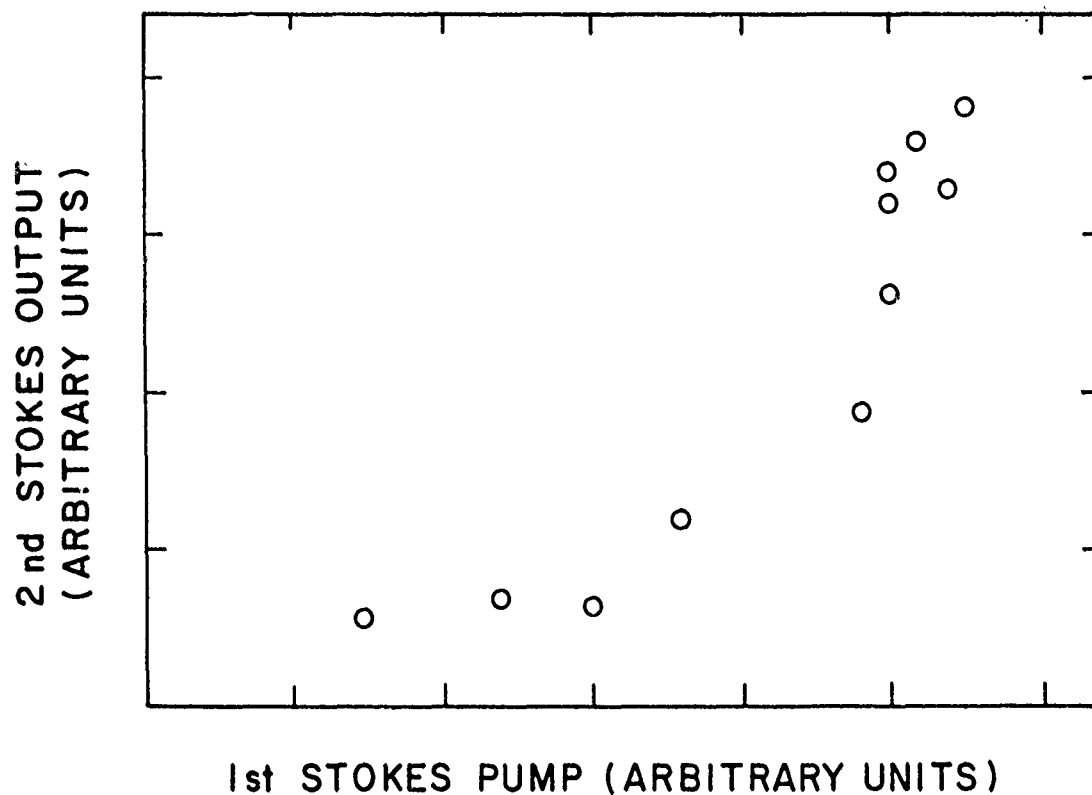
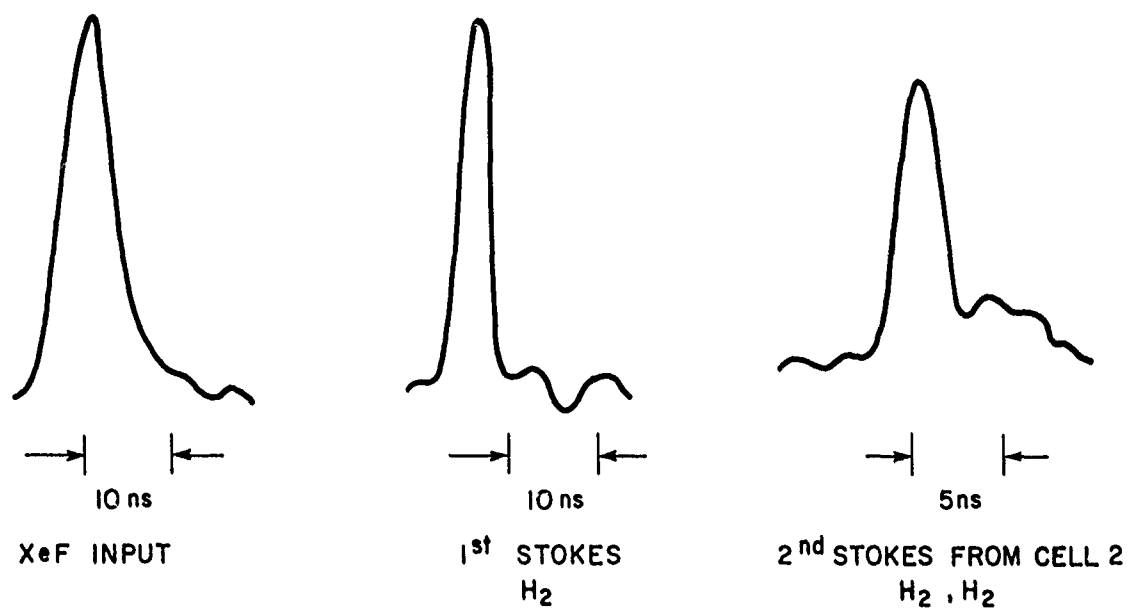


Figure 17 Cell 2 ( $H_2, H_2$ ) Pressure Variation Effect on S<sub>1</sub> Pump and the Generation of S<sub>2</sub>



J3724

Figure 18 Cell 2 Threshold for  $S_2$  as a Function of  $S_1$  Input Intensity



J3749

Figure 19 Pulse Temporal Narrowing in Successive Conversion

The typical energy conversion efficiency measured in this manner was 25 to 30% into  $S_2$  output from the  $S_1$  input, i.e., 495 out/410 in. This corresponds to a photon conversion efficiency of 30 to 35%, a power efficiency due to pulse shortening of 40 to 48% and a photon power efficiency of 48 to 56%. The total energy conversion efficiency from the XeF input into cell 1 into  $S_2$  output from cell 2 was 12% (34% photon power conversion). As discussed above, conversion efficiency characteristics of what was achieved in a single cell should be achievable in any subsequent cell for an overall conversion of  $\eta_1 \eta_2$ . Since we measured power conversion efficiencies of 66% in a single cell, overall conversion  $> 40\%$  to the blue-green should be achievable, but were unobtainable here due to the low pump intensities available.

This conjecture was tested somewhat by conducting two-step experiments with KrF\* as the pump. The KrF laser operates at higher output intensities and was capable of generating significant quantities of  $S_1$  through  $D_2$  (see Figure 20). In this case, the first cell contained 67 atm of  $D_2$  which resulted in an output consisting mainly of depleted pump (249 nm) and  $S_1$  at 268 nm. These laser transitions were collected and focussed into the second cell containing  $H_2$ . They each generated their own Stokes components in cell 2 but, most of the output consisted of a few first Stokes orders (see Figure 20). Of particular interest is the fact that the 268 nm  $S_1$  output of cell 1 was depleted over 70% in cell 2, with the majority of the output appearing in the sought after 308 nm line (which would be analogous to the production of 469 and 472 with XeF as the pump).

#### B. LONG PULSE EXPERIMENTS

Included as part of this DARPA-supported program, we also investigated Raman scattering using the output of a one-meter laser as the pump for single pass, single cell spectral observations. To perform these experiments, two different e-beam



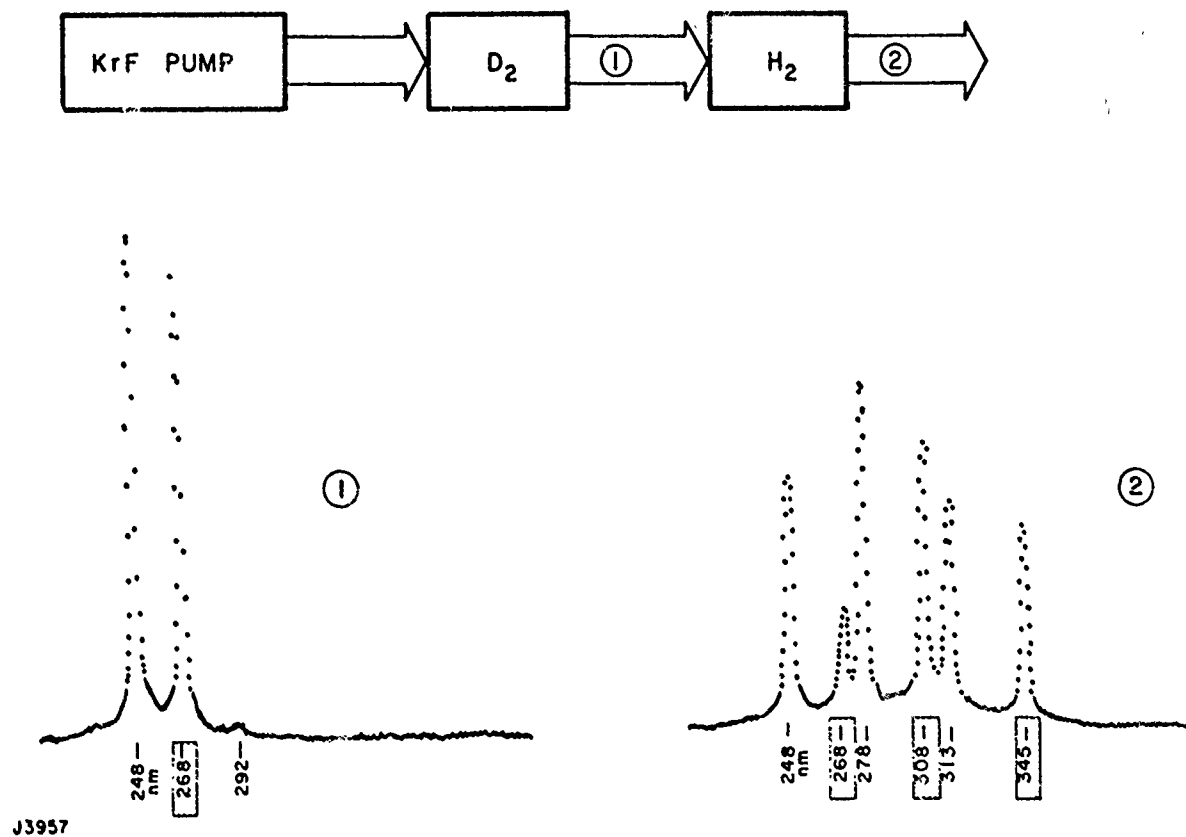


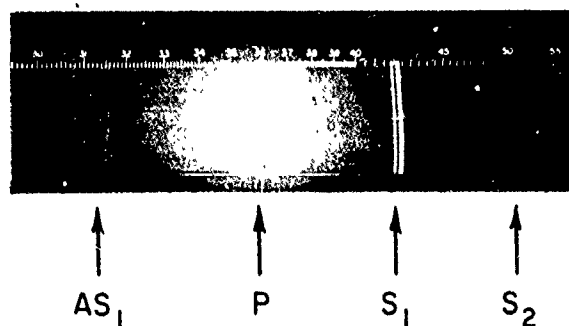
Figure 20 Two Step KrF/D<sub>2</sub>/H<sub>2</sub> Raman Conversion

laser devices were used. The first experiments which were carried out last summer involved the  $(10 \times 10 \times 100) \text{ cm}^3$  device constructed in support of the N00014-76-C-1032 DARPA-funded contract. It was run with conventional optics and provided  $\sim 5 \text{ J}$  in  $600 \text{ ns}$  for the few experiments we tried. Under these conditions, the output was collected by a 6-in. Ultrasil quartz plano-convex lens ( $50 \text{ cm fl}$ ) and focused into the center of our high pressure  $\text{H}_2$  cell ( $\sim 10 \text{ atm}$ ). We observed output spectra corresponding to  $S_1$ ,  $S_2$  and  $AS_1$  (see Figure 21). From color Polaroid open shutter photographs, it appeared that the output consisted of principally parametric four-wave conversion processes as was evidenced by the observation of annular rings. Also, the intensity correlations in the Stokes radiation ( $AS_1 > S_2$ ) supported this contention.

More recently, we performed SRS experiments using a  $(2 \times 2 \times 100) \text{ cm}^3$  e-beam device operating with  $\text{KrF}^*$  at  $249 \text{ nm}$ . This device produced  $\sim 0.5 \text{ J}$  in  $0.3 \mu\text{s}$  with flat (stable) optics. This output was again focussed into the high-pressure  $\text{H}_2$  cell ( $> 30 \text{ atm}$ ) and significant conversion to  $S_1$ ,  $S_2$  and some  $S_3$  was observed using the OMA (see Figure 22). Energy measurements showed energy conversion to these Stokes lines was significantly  $> 10\%$ . Also, photodiode measurements of the pump pulse and first Stokes pulse showed conversion occurred over the entire extended pulselength (see Figure 23). We believe the temporal features seen in the Raman output represent amplification of small-scale temporal structure in the laser pump due to the many cavity modes present in the stable optics (flat-flat) setup.

This cavity has been modified to include Brewster windows and unstable optics (see Figure 24). Energy output near  $0.6 \text{ J}$  in  $0.4 \text{ s}$  has been routinely observed. Using this beam, we have observed long pulselength  $\text{XeF}^*$  SRS in  $\text{H}_2$  (see Figure 25). Here the OMA shows conversion is principally into  $S_1$  ( $413 \text{ nm}$ ) with significant depletion of the  $\text{XeF}$  pump and generation of  $S_2$  ( $499 \text{ nm}$ ). These data are not calibrated and these

LONG PULSE LENGTH  $\text{XeF}^*$  SRS EXPERIMENT  
(10 ATM  $\text{H}_2$ )



J3827

Figure 21 Spectra Giving Qualitative Indication of Relative Intensities of  $\text{XeF}$  Pumped  $\text{H}_2$  ( $\text{AS}_1$ , P,  $\text{S}_1$ ,  $\text{S}_2$ )

# LONG PULSE LENGTH KrF\* SRS EXPERIMENT (H<sub>2</sub>)

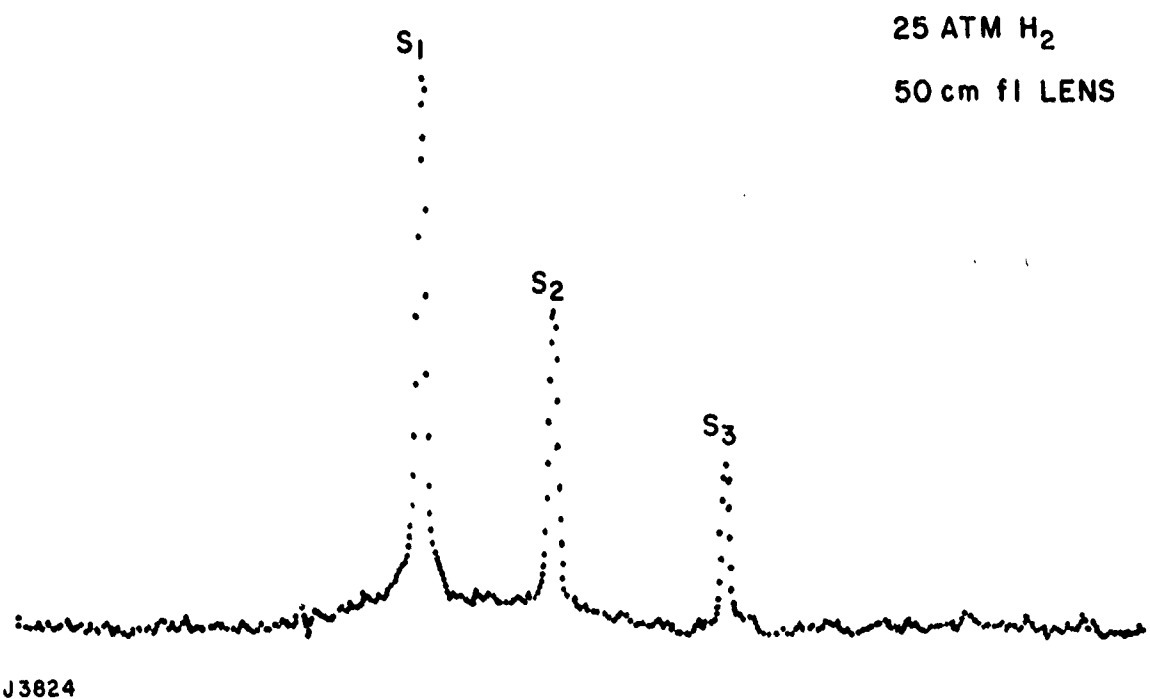
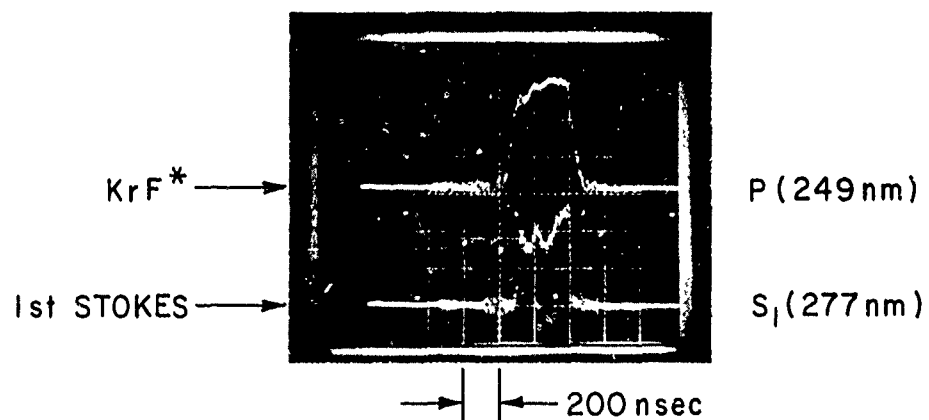


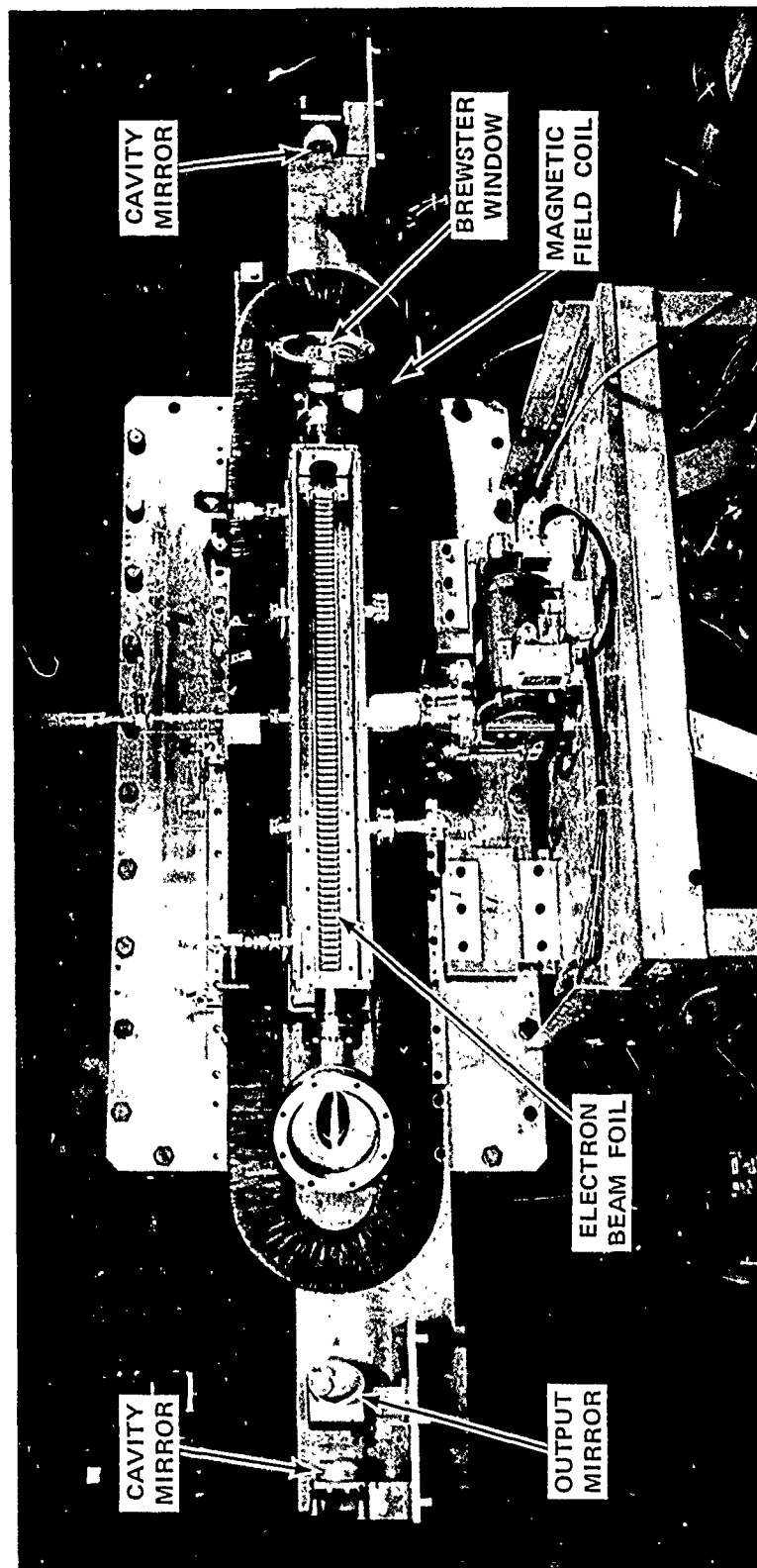
Figure 22 OMA Output for KrF Pumped H<sub>2</sub> with Filter to Remove Pump (249 nm) and Shorter Wavelengths

# KrF\* PUMPED SRS EXPERIMENT (H<sub>2</sub>)



J3828

Figure 23 Temporal Pulse Shapes of Input KrF Pump and S<sub>1</sub> Stokes Shifted Raman Pulse



J3829

Figure 24 Photograph of Modified One Meter Laser

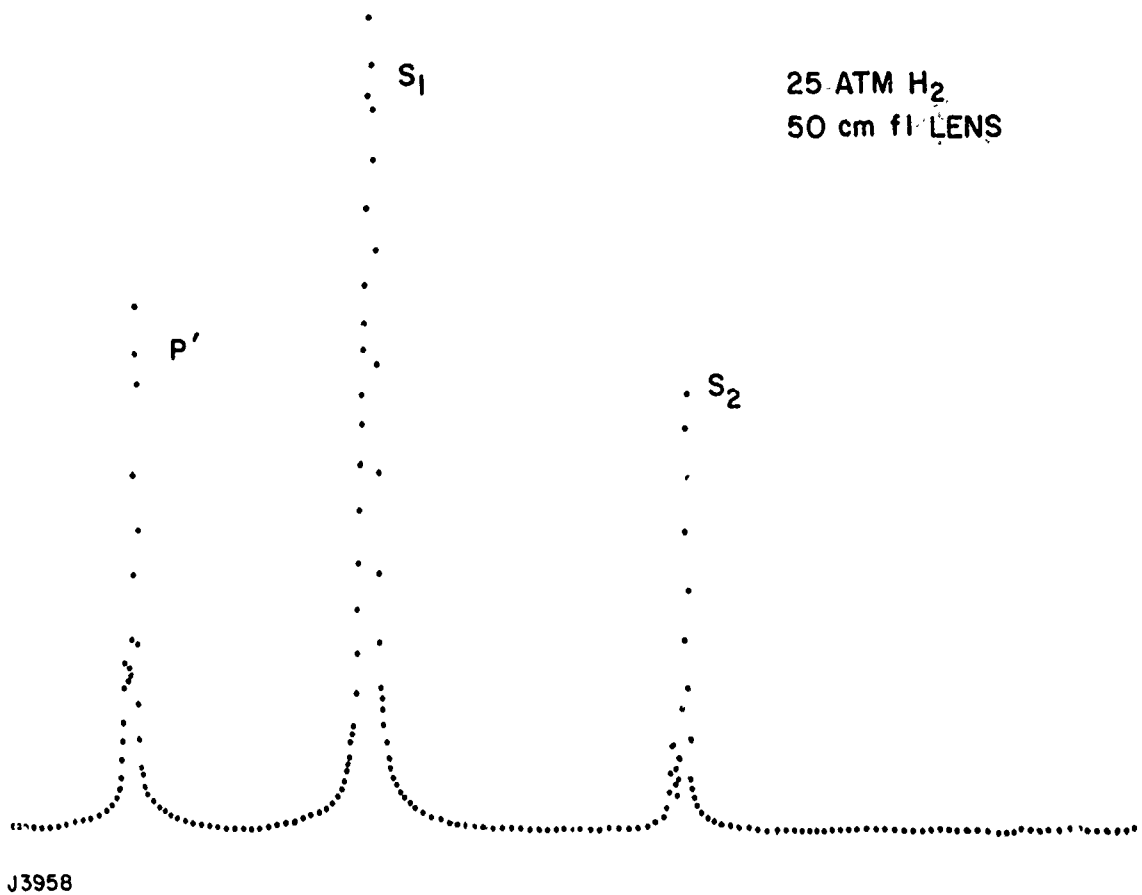
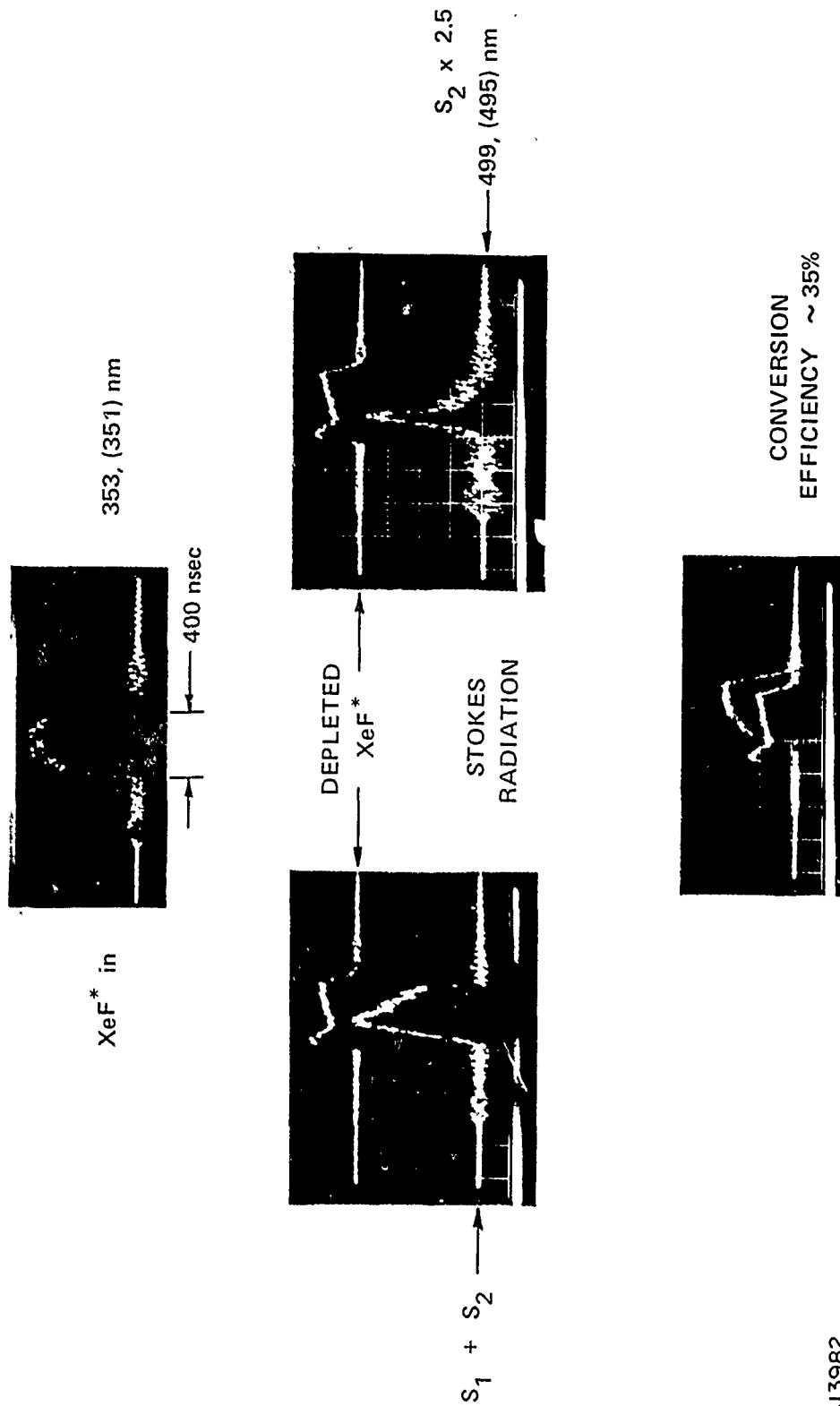


Figure 25 Long Pulse Length XeF\* SRS Experiment ( $H_2$ )

spectral results are merely qualitative. More quantitative results are shown in Figure 26 where the XeF pump was softly focused ( $\sim 135$  cm) into about 30 atm  $H_2$ . Data collected with photodiodes and appropriate filters showed  $\sim 35\%$  pump depletion with the majority going into first stokes in good agreement with our expectations. These conversion experimental results are preliminary. Further experimentation is underway and will be summarized in subsequent Interim Technical Reports.





J3982

Figure 26 XeF\* Pumped SRS Experiment (H<sub>2</sub>) (One Meter Device)

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